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LEVEL II

DYE LASER PARAMETER CALCULATIONS

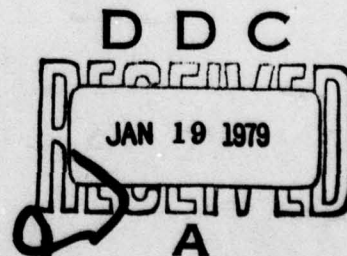by

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March 1976

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NAVAL WEAPONS CENTER

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FOREWORD

This report has been prepared for the timely presentation of information on calculational procedures for dye laser parameters and is released at the working level. The methods used here are fairly general and can be applied to other laser systems as well. Further, the report provides an intuitive basis for reasoning about laser phenomena.

This research was performed for the period July 1974 through June 1975 and was supported by Naval Sea Systems Command under Task No. F32344406. This is an interim report and more detailed calculational procedures will be published as the work continues.

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Head, Physics Division
Research Department
22 July 1975

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15. OUTLINE, TABLE OF CONTENTS, SUMMARY, OR EQUIVALENT DESCRIPTION

This report has been prepared for the timely presentation of information on calculational procedures for dye laser parameters and is released at the working level. The methods used here are fairly general and can be applied to other laser systems as well. Further, the report provides an intuitive basis for reasoning about laser phenomena.

Although several textbooks give elementary treatments to calculations of laser thresholds, power output, and gain saturation, none of these is really adequate for the specific case of dye lasers for two reasons: (1) the textbooks calculate thresholds in terms of spontaneous lifetimes, which in dyes are very short and hard to measure, and (2) the textbooks fail to give a "feel" for the microscopic atomic events which occur in the laser because they perform the entire calculation in terms of macroscopic quantities. Therefore, for this study it was decided to treat the laser problem in terms of experimentally measurable atomic emission and absorption cross section.

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16. KEY WORDS FOR INDEXING
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INTRODUCTION

Although several textbooks[1-3] give elementary treatments to calculations of laser thresholds, power output, and gain saturation, none of these is really adequate for the specific case of dye lasers for two reasons: (1) the textbooks calculate thresholds in terms of spontaneous lifetimes, which in dyes are very short and hard to measure, and (2) the textbooks fail to give a "feel" for the microscopic atomic events which occur in the laser because they perform the entire calculation in terms of macroscopic quantities. Therefore, for this study it was decided to treat the laser problem in terms of experimentally measurable atomic emission and absorption cross sections.

LASER DYE ENERGY LEVELS AND SPECTRAL PROPERTIES

Figure 1 is an energy level diagram of a dye molecule. Lasing occurs between the singlet S_1 and S_0 electronic states. Within both S_1 and S_0 is a subset of vibrational and rotational energy levels. The transition lifetime from S_1 to T_1, the lowest triplet level, is usually several hundred nanoseconds so that for pumping pulses of less than, say 200 ns, we need not consider the triplet population buildup. Also, the absorption at the lasing wavelength due to $S_1 \rightarrow S_2$ transitions is neglected here because examples[4,5] tend to indicate that the $S_1 \rightarrow S_2$ absorption peak is farther removed from the $S_1 \rightarrow S_0$ fluorescence peak than is the $S_0 \rightarrow S_1$ absorption peak. Figure 2 shows the absorption fluorescence spectra of rhodamine B; the obvious "mirror" symmetry of the spectra is a result of configuration coordinate conservation during transitions as

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[1] Yariv, A. *Introduction to Optical Electronics*. New York, Holt, Rinehart, and Winston, 1971.

[2] Bass, M., T. F. Deutsch, and M. J. Weber. "Dye Lasers," in *Lasers*, ed. by A. K. Levine. New York, Marcel Dekker, Inc., 1971. Vol. 3, p. 287.

[3] Lengyel, B. A. *Lasers*. New York, John Wiley and Sons, 1971.

[4] Bonneau, R., J. Faure, and J. Joussot-Dubien. "Singlet-Singlet Absorption and Intersystem Crossing From the $^1B_{3u}$ State of Napthalene," CHEM PHYS LETT, Vol. 2 (June 1968), p. 65.

[5] Pavlopoulos, T. G. "Prediction of Laser Action Properties of Organic Dyes From Their Structure and the Polarization Characteristics of Their Electronic Transitions," IEEE J QUANTUM ELECTRON, Vol. QE-9 (June 1973), p. 510.

detailed in the Franck-Condon principle. The peaks of the absorption
and fluorescence curves are separated by a few hundred angstroms and the
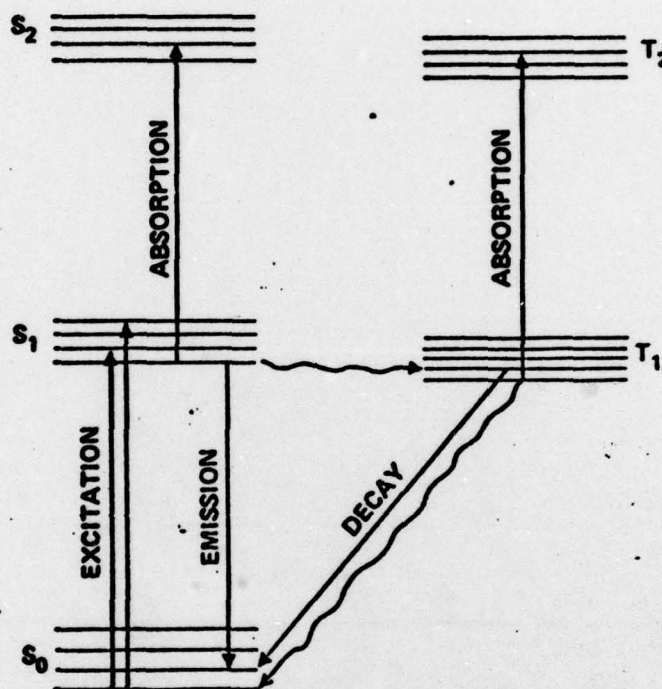fluorescence peak is always at a longer wavelength than the absorption
peak.

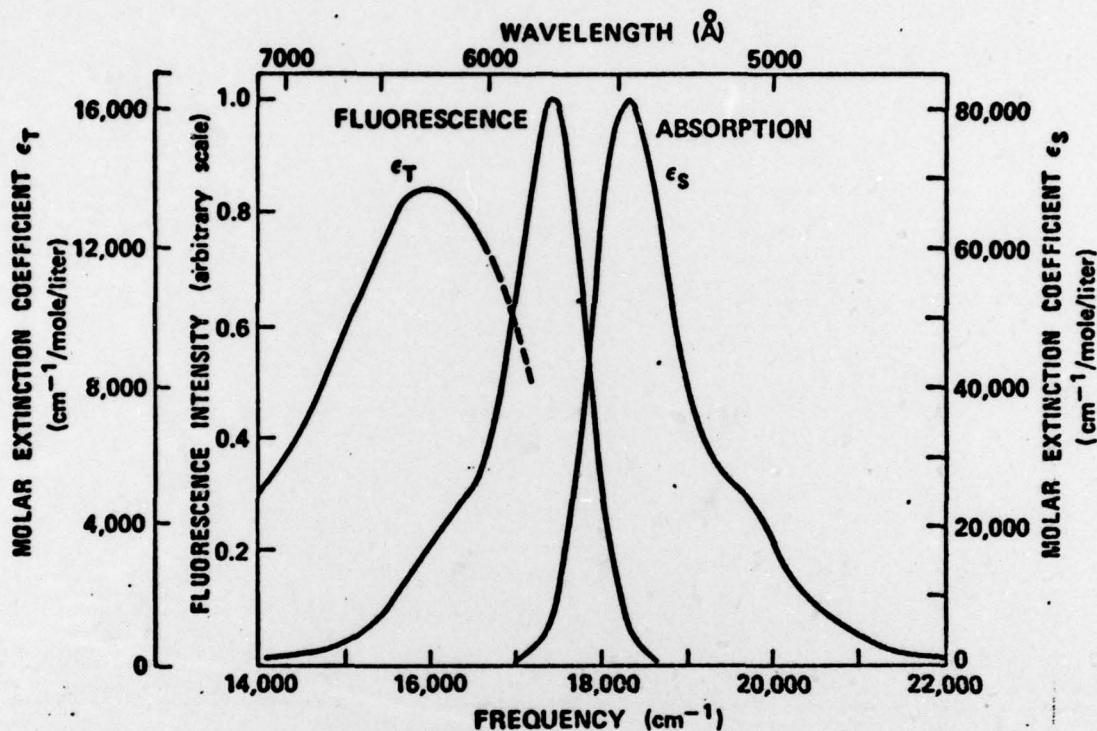FIGURE 1.   Energy Level Scheme of a
Dye Molecule.

FIGURE 2. Singlet Absorption and Fluorescence Spectra.

MATHEMATICAL TERMINOLOGY

We will use microscopic quantities such as atomic cross sections and photon flux densities throughout the main body of calculations until, at the end, we will convert these to their macroscopic analogues. We therefore briefly review the definitions of the microscopic quantities.

If a photon flux density, ϕ (photons$\cdot$cm$^{-2}\cdot$s^{-1}), is incident on a system of atoms of density n (atoms$\cdot$cm^{-3}) and interaction cross section σ (cm^2) then the number of interactions taking place per second in 1 cm^3 of these atoms is n$\sigma\phi$ (interactions$\cdot$cm$^{-3}\cdot$s^{-1}). This defines the interaction cross section, σ. In general, however, neither ϕ nor σ will be constant functions of photon frequency, ν, and we therefore define n$\sigma(\nu)\phi(\nu)\Delta\nu$ to be the number of interactions in 1 cm^3 in 1 second where $\phi(\nu)$ is the photon flux density Hz^{-1} and $\Delta\nu$(Hz) is the frequency bandwidth associated with either $\sigma(\nu)$ or $\phi(\nu)$ or a convolution of both. Note that nσ = α, the absorption rate, (cm^{-1}) in the medium. The effective absorption rate due to the finite transmission T of the end mirrors

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is $\alpha_M = -\ln(R_1 R_2)/(2\ell)$ (cm^{-1}) where ℓ is the length of the active medium and $R = (1 - T)$. In a dye laser, both dye flow turbulence and/ or heating of the dye medium give rise to refraction out of the beam, and scattering and diffraction also cause additional losses so that we group the mirror loss, α_M, and other losses, α_R, together to form a term $\alpha_{loss} = \alpha_M + \alpha_R$. The values for σ may be obtained from the experimentally measured absorption and stimulated emission curves, $\sigma(\nu)$. Because the density of states for upward and downward transitions is usually about the same, we expect that $\sigma_{10}(\nu)$ for stimulated emission at its peak wavelength will be roughly equal to $\sigma_{01}(\nu)$ for absorption at the peak absorption wavelength.

LASING THRESHOLD CALCULATIONS

During steady state pumping, with no optical wave in the cavity, and without considering the effects of cavity blackbody radiation, the pump-induced upward transitions per cm^3-s are just equal to the number of spontaneously induced downward transitions per cm^3-s, both radiative and nonradiative. The *radiative* spontaneous transitions are induced by the effective zero point photon flux density, $\phi_0 = 8\pi n_d^2 \nu^2/c^2$ (photons cm^{-2} s^{-1}-Hz^{-1}), where n_d = refractive index of dye medium, ν = photon frequency, and c = speed of light in vacuum. Appendix A shows that this effective flux does indeed give the correct spontaneous decay rate when applied to a dipolar interaction cross section. ϕ_0 is a continuous function of ν, and it acts on the stimulated emission band of width $\Delta\nu_f$, peaked at ν_f, where $\Delta\nu_f$ is the full width at half maximum of the fluorescence spectral emission intensity curve. Alternately,

$$\Delta\nu_f = \int_0^\infty \sigma_{10}(\nu)\phi_0(\nu)d\nu/[\sigma_{10}(\nu_f)\phi_0(\nu_f)]$$

where σ_{10} is the emission cross section and ν_f is the frequency at which $\sigma_{10}\phi_0$ is maximum. Thus, in steady state pumping, with no optical wave (i.e., below threshold):

$$\phi_{in}(\nu_p)n_0\sigma_{01}(\nu_p)\Delta\nu_p = \phi_0(\nu_f)n_1\sigma_{10}(\nu_f)\Delta\nu_f/\eta \qquad (1)$$

where subscripts 0 and 1 refer to the S_0 and S_1 singlet electronic states of Figure 1; η, the fluorescence efficiency, corrects for nonradiative transitions from S_1 to S_0; and $\Delta\nu_p$ is the spectral linewidth in hertz of the pump input, ϕ_{in}, centered at ν_p. Now $\alpha(\nu_g)$, the average gain per centimeter that a photon flux of frequency ν_g experiences in the active medium, is

$$\alpha(\nu_g) = n_1\sigma_{10}(\nu_g) - n_0\sigma_{01}(\nu_g) - \alpha_{loss} \qquad (2a)$$

$\alpha(\nu_g)$ must be zero during steady state lasing. Thus

$$n_1 = \frac{n_0\sigma_{01}(\nu_g) + \alpha_{loss}}{\sigma_{10}(\nu_g)} \tag{2b}$$

Combining Equation 1 and 2b we obtain:

$$\phi_{in}^{threshold} = \frac{\phi_0\sigma_{10}(\nu_f)}{\eta\sigma_{01}(\nu_p)} \frac{\sigma_{01}(\nu_g)}{\sigma_{10}(\nu_g)} \frac{\Delta\nu_f}{\Delta\nu_p}\left(1 + \frac{\alpha_{loss}}{n_0\sigma_{01}(\nu_g)}\right)$$

or in terms of irradiance, H (W cm^{-2})

$$H_{in}^{thr}(\nu_g) = h\nu_p\Delta\nu_p\phi_{in}^{thr} = \frac{8\pi h n_d^2\nu_f^2\nu_p\Delta\nu_f}{nc^2}\frac{\sigma_{10}(\nu_f)}{\sigma_{01}(\nu_p)}\frac{\sigma_{01}(\nu_g)}{\sigma_{10}(\nu_g)}\left(1 + \frac{\alpha_{loss}}{n_0\sigma_{01}(\nu_g)}\right)$$

where h is Planck's constant. In terms of wavelength, λ:

$$H_{in}^{thr}(\lambda_g) = \frac{8\pi h n_d^2 c^2\Delta\lambda_f}{n\lambda_f^4\lambda_p}\frac{\sigma_{10}(\lambda_f)}{\sigma_{01}(\lambda_p)}\frac{\sigma_{01}(\lambda_g)}{\lambda_{10}(\lambda_g)}\left(1 + \frac{\alpha_{loss}}{n_0\sigma_{01}(\lambda_g)}\right) \tag{2c}$$

This is the irradiance level required to transverse-pump the portion of the dye immediately adjacent to the dye-pump window interface to threshold. In order to obtain a threshold inversion to depth d by transverse-pumping it is necessary to increase this irradiance by the factor $\exp[\alpha(\nu_p)d]$. Appendix B describes a method of measurement of $\sigma_{10}(\nu)$.

Threshold numerical example: For the dye emission and absorption curves illustrated in Figure 3 we have

$\lambda_f = 0.95$ μm

$\lambda_p = 0.82$ μm

$\Delta\lambda_f = 0.10$ μm

$h = 6.6 \times 10^{-34}$ joule·s

$n_d = 1.4$

$\eta = 0.2$

Set $\sigma_{10}(\lambda_f)/\sigma_{01}(\lambda_p) = 1$.

Thus, $H_{in} = 2.2 \times 10^9 [\sigma_{01}(\lambda_g)/\sigma_{10}(\lambda_g)][1 + \alpha_{loss}/n_0\sigma_{01}(\lambda_g)]$W·m^{-2}.

Typically $\sigma_{01}(\nu_g)/\sigma_{10}(\nu_g) \lesssim 0.01$ as in Figure 3 so that if

$\alpha_{loss} \ll n_0\sigma_{01}(\lambda_g)$, $H_{in} \approx 2.2 \times 10^3$ W·cm^{-2}. This corresponds to the input irradiance needed to invert the dye medium sufficiently to counteract its own absorption.
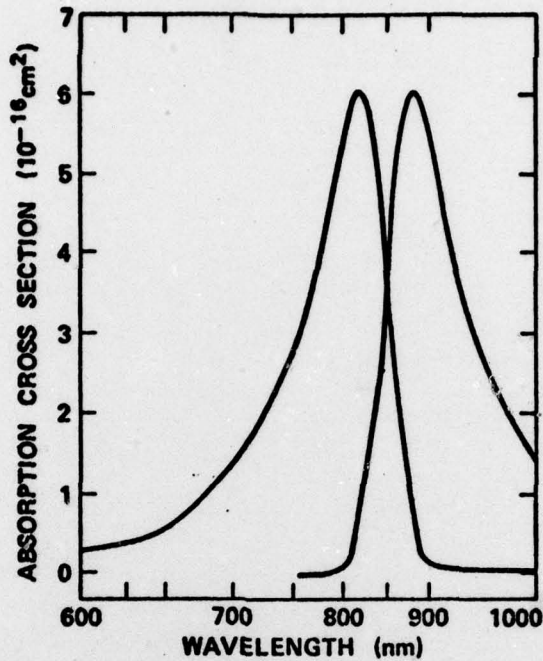
FIGURE 3. Dye Emission and Absorption
Cross Section Versus Wavelength.

GAIN SATURATION IN HOMOGENEOUS DYE LASER MEDIA

Here we use an equation for steady state operation similar to
Equation 1, but now we are above threshold so we must include the upward
and downward transitions due to the optical photon flux density ϕ_{opt}.
(Again we neglect $S_1 \rightarrow S_2$ and $T_1 \rightarrow T_2$ transitions.)

$$n_0\phi_{in}(\nu_p)\sigma_{01}(\nu_p)\Delta\nu_p + n_0\phi_{opt}(\nu_g)\sigma_{01}(\nu_g)\Delta\nu_g = (n_1/n)\phi_0(\nu_f)\sigma_{10}(\nu_f)\Delta\nu_f$$

$$+ n_1\phi_{opt}(\nu_g)\sigma_{10}(\nu_g)\Delta\nu_g \tag{3}$$

The gain in the laser media is:

$$\alpha(\nu_g) = n_1\sigma_{10}(\nu_g) - n_0\sigma_{01}(\nu_g) \tag{4}$$

Combining Equation 3 and 4 leads to

$$\alpha(\nu_g) = \frac{n n_0 [\sigma_{10}(\nu_g)\phi_{in}(\nu_p)\sigma_{01}(\nu_p)\Delta\nu_p - \sigma_{01}(\nu_g)\phi_0(\nu_f)\sigma_{10}(\nu_f)\Delta\nu_f/\eta]}{\phi_0(\nu_f)\sigma_{10}(\nu_f)\Delta\nu_f \left[1 + \dfrac{\eta\phi_{opt}\sigma_{10}(\nu_g)\Delta\nu_g}{\phi_0(\nu_f)\sigma_{10}(\nu_f)\Delta\nu_f}\right]} \tag{5}$$

so that the one-half gain point for α is reached when the second term in brackets in the denominator is equal to one or:

$$\phi_{opt}^{sat} = \frac{\sigma_{10}(\nu_f)}{\sigma_{10}(\nu_g)} \frac{\Delta\nu_f}{\Delta\nu_g} \frac{8\pi n_d^2 \nu_f^2}{c^2 \eta}$$

It has been shown that even on the assumption that the dye fluorescence band is composed of many narrow homogeneous spectral lines, due to very rapid spectral diffusion of the lower energy levels of the dye in its solvent, the gain saturates at the same input flux density as given above.[6]

DYE LASER OUTPUT POWER AND EFFICIENCY

During steady state lasing the average gain in the cavity is zero so that again, just as at threshold, we have:

$$n_1 = [n_0\sigma_{01}(\nu_g) + \alpha_{loss}]/\sigma_{10}(\nu_g) \tag{2b}$$

Using Equation 3 with 2b we obtain

$$\alpha_{loss}(\Delta\nu_g)\phi_{opt} = n_0\phi_{in}(\nu_p)\sigma_{01}(\nu_p)\Delta\nu_p - n_0\sigma_{01}(\nu_g)\phi_0\sigma_{10}(\nu_f)\Delta\nu_f/[\sigma_{10}(\nu_g)\eta]$$

$$- \phi_0(\nu_f)\sigma_{10}(\nu_f)\Delta\nu_f\alpha_{loss}/[\sigma_{10}(\nu_g)\eta]$$

Using our former calculation to identify ϕ_{in}^{thr} we obtain:

$$\phi_{out} = \phi_{opt}T = \frac{T\phi_{in}n_0\sigma_{01}(\nu_0)\Delta\nu_p}{\alpha_{loss}\Delta\nu_g}\left[1 - \frac{\phi_{in}^{thr}}{\phi_{in}}\right] \tag{6}$$

Now $H = h\nu\phi\Delta\nu$.

$$H_{out} = H_{in}\frac{\nu_g T n_0\sigma_{01}(\nu_p)}{\nu_p\alpha_{loss}}\left[1 - \frac{\phi_{in}^{thr}}{\phi_{in}}\right]$$

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[6] Mourou, Gerald. "Spectral Hole Burning in Dye Solutions," IEEE J QUANTUM ELECTRON, Vol. QE-11 (January 1975), pp. 1-8.

Now for transverse-pumping the input power, $P_{in} = H_{in}\ell h$ where ℓ and h are length and height of pumped region. Then $P_{out} = H_{out}h/n_0\sigma_{01}(\nu_p)$ where $[n_0\sigma_{01}(\nu_p)]^{-1}$ is the depth of the pumped region so that

$$P_{out} = \frac{P_{in}T(\nu_g/\nu_p)}{\ell\,\alpha_{loss}}\left[1 - \frac{\phi_{in}^{thr}}{\phi_{in}}\right]$$

In the limit that $T \ll 1$ we have $\alpha_M \approx T/\ell$ so that

$$P_{out} \approx P_{in}\nu_g/\nu_p[1 - \phi_{in}^{thr}/\phi_{in}][1/(1 + \alpha_R\ell/T)]$$

Note that this is independent of η, the fluorescence efficiency, if $\phi_{in} \gg \phi_{in}^{thr}$. The power efficiency, P_{out}/P_{in}, when far above threshold is usually limited by the $\alpha_R\ell/T$ term in the denominator because the other losses usually exceed the useful output.

NOISE IN DYE LASER AMPLIFIERS[7]

This noise is just due to the amplified spontaneous emission into the acceptance solid angle, $d\Omega$, and the band pass, $d\nu$, of the amplifier. The number of spontaneous interactions per unit volume in this category is:

$$\frac{dN}{dV} = \int n_1\phi_0(\nu_f)\sigma_{10}(\nu_f)d\nu\,\frac{d\Omega}{4\pi}$$

Now if our amplifier length is ℓ (cm) and has cross sectional area A and exponential gain constant α, then the photon noise output is:

$$N = A\int_0^\ell dz\,\frac{dN}{dV}\,e^{\alpha(\ell-z)}$$

$$= A\,\frac{(e^{\alpha\ell} - 1)}{\alpha}\,\frac{dN}{dV}$$

Now $\alpha = n_1\sigma_{10}(\nu_g) - n_0\sigma_{01}(\nu_g)$ and if the signal photon flux is much less than the saturation flux, then in steady state

$$n_0\phi_{in}(\nu_p)\sigma_{01}(\nu_p)\Delta\nu_p = n_1\phi_0(\nu_f)\sigma_{10}(\nu_f)\Delta\nu_f/\eta$$

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[7] Yariv, A. *Quantum Electronics*. New York, John Wiley and Sons, 1967. P. 412.

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so that

$$\alpha = n_1[\sigma_{10}(\nu_g) - \sigma_{01}(\nu_g)\phi_0(\nu_f)\sigma_{10}(\nu_f)\Delta\nu_f/(\eta\phi_{in}\sigma_{01}(\nu_\rho)\Delta\nu_p)]$$

or

$$\alpha = n_1\sigma_{10}(\nu_g)(1 - \phi_{in}^{crit}/\phi_{in})$$

Then

$$N/A = \int(e^{\alpha\ell} - 1)\frac{[\phi_0(\nu_f)d\nu d\Omega/4\pi][\sigma_{10}(\nu_f)/\sigma_{10}(\nu_g)]}{(1 - \phi_{in}^{crit}/\phi_{in})}$$

where ϕ_{in}^{crit} is the input flux density necessary to cause zero exponential gain in the medium. Referred to the input of the amplifier, the amplifier noise is

$$(N/A)_{input} = \frac{(e^{\alpha\ell} - 1)2(\nu^2/c^2)\Delta\nu\Delta\Omega}{e^{\alpha\ell}(1 - \phi_{in}^{crit}/\phi_{in})}$$

Numerical example: For a typical amplifier for an optical system we choose a 3-degree field of view, $\alpha\ell = 100$, $\phi_{in} \gg \phi_{in}^{crit}$, $\Delta\nu = 10^8$ Hz (to allow for Doppler), $\nu/c = 1/\lambda = 10^4 \cdot cm^{-1}$

$$\Delta\Omega = \pi(3/57.3)^2 = 0.0086 \text{ Sr}$$

$$(N/A)_{input} = 2 \cdot 10^8 \cdot (0.0086) \cdot 10^8 = 1.7 \times 10^{14} \text{ photons} \cdot cm^{-2} \cdot s^{-1}$$

$$(NEI)_{steady} = (h\nu)(N/A)_{input} = (6.6 \times 10^{-34})(3 \times 10^{14})(1.7 \times 10^{14})$$

$$= 3.4 \times 10^{-5} \text{ W} \cdot cm^{-2}$$

where $(NEI)_{steady}$ = steady state noise equivalent irradiance with a 10^8 Hz optical filter at the output of the laser amplifier, before the detector. If, instead, we want to know the NEI for *fluctuations* about the steady state irradiance, then it can be shown[8] that the noise/$\sqrt{Hz}$ is $\sqrt{(N/A)_{input}}$ or 1.3×10^7 photons $\cdot cm^{-1} \cdot s^{-1/2}$ and with B, the detector bandwidth, equal to 10^8 Hz we have

$$(NEI)_{fluct} = h\nu\sqrt{(N/A)}\sqrt{B}$$

$$= (6.6 \times 10^{-34})(3 \times 10^{14})(1.3 \times 10^7)(10^4)$$

$$= 2.57 \times 10^{-8} \text{ W} \cdot cm^{-2}$$

[8] Freed, C., and H. A. Haus. "Photoelectron Statistics Produced by a Laser Operating Below and Above the Threshold of Oscillation," IEEE J QUANTUM ELECTRON, Vol. QE-2 (August 1966), p. 190.

For a commonly used simple silicon detector of 1 cm^2 area at 1 μm the noise equivalent power is 1×10^{-12} W·Hz$^{-1/2}$ or, for the above 10^8 Hz bandwidth, the NEI for a system without optics using this detector is $(\sqrt{10^8}) \times (1 \times 10^{-12}) = 10^{-8}$ W·cm^{-2}. If we use this detector with a 3-degree field of view f/1 optical system, then the objective diameter is 19.1 cm and the NEI for this system becomes 3.5×10^{-11} W·cm^{-2}, which is much lower than our $(NEI)_{fluct}$ of our dye amplifier.

Note that decreasing the angular field of view requirement allows decreasing both the acceptance angle, $\Delta\Omega$, of our laser amplifier and the size of the detector in the direct detection system. These effects cancel so that the ratio of NEI's remains constant, down to a detector size limited by the noise of available preamplifiers.

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Appendix A

EFFECTIVE ZERO POINT PHOTON FLUX DENSITY, ϕ_0

We wish to show that the spontaneous transition rate is related through the interaction cross section σ_{10} of the atom to an effective zero point photon flux density $\phi_0 = (8\pi n_a^2 \nu^2/c^2)$. To do this we must calculate the time dependence of the state coefficients for a perturbation caused by this zero field or vacuum photon flux density. The Hamiltonian for a two-level, a and b, atomic system and a photon field and the interaction thereof is given by[9]

$$H = \sum_s \hbar\Omega\left(a_s^+ a_s + 1/2\right) + \hbar\begin{pmatrix} \omega_a & 0 \\ 0 & \omega_b \end{pmatrix} + \hbar\sum_s g_s\left(\sigma a_s^+ + \sigma^+ a_s\right)$$

Here a_s^+ and a_s are the photon creation and annihilation operators, respectively; $\hbar\omega_a$ and $\hbar\omega_b$ are the energies of atomic levels a and b, respectively. σ and σ^+ are the atomic raising and lowering matrices, while g_s is the electromagnetic field-atom coupling constant where $g_s^2(\Omega) = \mu^2\Omega/\hbar V\varepsilon_0(\sin^2 Kz)$. The quantities of which g_s is composed are defined below:

μ = electric dipole matrix element between levels a and b
Ω = frequency of photons (assumed monochromatic at present (rad s^{-1})
V = volume considered
ε_0 = permeability of vacuum = 8.85×10^{-12} F·m^{-1}
K = wave number of the radiation field

Now the exponential atomic decay rate due to this generalized perturbation (which includes the zero point fluctuations)[6] is:

$$\gamma = 2\pi g^2(\omega)D(\omega) \quad (s^{-1})$$

where $D(\omega) = V\omega^2/\pi^2 c^3$. Thus

$$\gamma = 2\pi \frac{\mu^2}{\hbar\pi^2} \frac{\omega^3}{c^3\varepsilon_0} \sin^2 Kz = \frac{16\pi^2\mu^2\nu^3}{\hbar c^3\varepsilon_0} \sin^2 Kz$$

[9] Sargent, M., M. O. Scully, and W. E. Lamb. *Laser Physics.* Reading, Mass., Addison-Wesley Publishing Co., 1974. P. 237.

where $\nu = \omega/2\pi$. Now we wish to average over all z and to convert the charge units in μ^2 to gaussian units. This may be done by multiplying by $4\pi\epsilon_0/2$ and dropping the $\sin^2 Kz$. Then

$$\gamma = \frac{32\pi^3\mu^2\nu^3}{\hbar c^3}$$

Eisberg[10] gives a decay rate for a monochromatic collimated input wave $2E_x\cos 2\pi\nu t$:

$$\gamma_{coll} = E_x^2\mu^2/(\hbar^2\Delta\nu) \quad \text{(gaussian units)}$$

where $\Delta\nu$ is the spectral linewidth of the transition. Now using Poynting's theorem

$$\phi_{coll} = c/(4\pi)\left[\frac{\langle E^2\rangle}{h\nu\Delta\nu}\right]$$

so that taking the average of E

$$\phi_{coll} = 2c/(4\ )\frac{E_x^2}{h\nu\Delta\nu}$$

Now $\gamma_{coll} = \sigma_{10}\phi_{coll}\Delta\nu$ or $\sigma_{10} = (\gamma_{coll}/\phi_{coll})/\Delta\nu$. Then

$$\sigma_{10} = \frac{E_x^2\mu^2 4\pi h\nu(\Delta\nu)}{2\hbar^2 cE_x^2(\Delta\nu)^2} = \frac{2\pi\mu^2 h\nu}{\Delta\nu\hbar^2 c}$$

Then the effective value of photon flux density, $\phi_0\Delta\nu$, which "causes" the spontaneous transitions is just

$$\phi_0\Delta\nu = \gamma/\sigma_{10}$$

$$\phi_0\Delta\nu = \frac{32\pi^3\mu^2\nu^3}{\hbar^2 c^3}\frac{\hbar^2 c\Delta\nu}{2\pi\mu^2 h\nu}$$

$$\phi_0\Delta\nu = \frac{16\pi^2\nu^2}{c^2}\frac{\hbar\Delta\nu}{h} = (8\pi\nu^2/c^2)\Delta\nu \quad \text{(photons cm}^{-2}\text{ s}^{-1}\text{)}$$

or

$$\phi_0 = 8\pi\nu^2/c^2 \quad \text{(photons cm}^{-2}\cdot\text{s}^{-1}\text{ Hz}^{-1}\text{)}$$

---

[10] Eisberg, R. M. *Fundamentals of Modern Physics.* New York, John Wiley and Sons, 1961. P. 456.

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This is the effective radiative-spontaneous-emission-causing photon flux. In a dielectric medium the function $D(\omega)$ becomes $n_d^2 V \omega^2 / \pi^2 c^3$, where n_d is the refractive index, and therefore ϕ_0 becomes

$$\phi_0 = 8\pi n_d^2 \nu^2 / c^2 \quad (\text{photons } cm^{-2} \cdot s^{-1} \cdot Hz^{-1})$$

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Appendix B

MEASUREMENT OF $\sigma_{10}(\nu)$

From Appendix A we know that:

$$\sigma_{10}(\nu_f)\phi_0(\nu_f)\Delta\nu_f = \int_0^\infty \sigma_{10}(\nu)\phi_0(\nu)d\nu = \tau_{rad}^{-1}$$

But τ_{rad} may be obtained from independent measurements of the lifetimes of the excited state and the fluorescence efficiency η, because

$$\tau_{rad} = \tau_{spontaneous}\,\eta^{-1}$$

$\tau_{spontaneous}$ is measured by observing the fluorescence decay after a fast fall time excitation pulse (a pulse from a mode locked laser, for example). For a good laser dye, this decay time will be on the order of 1 to 10 nanoseconds so it will be measurable on an oscilloscope.

The fluorescence efficiency, η, may be measured by Vavilov's method[11] which involves comparison of the isotropic light output of the fluorescent medium with the Lambertian scattering of the excitation source from a white scatterer such as MgO. Having obtained $\tau_{spontaneous}/\eta$ we then have a value

$$\int_0^\infty \sigma_{10}(\nu)\phi_0(\nu)d\nu = 1/\tau_{rad}$$

To get $\sigma_{10}(\nu)$ from this we must measure the relative curve of fluorescence intensity, $F(\nu)$, versus frequency and integrate. At any frequency, ν, the fluorescence intensity is proportional to $\sigma_{10}(\nu)\phi_0(\nu)$ so that the fluorescence width,

$$\Delta\nu_f = \int_0^\infty F(\nu)d\nu F_{max}^{-1} ,$$

of $F(\nu)$ is the same as the line width of the function $\sigma_{10}(\nu)\phi_0(\nu)$.

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[11] Demas, J. N., and G. A. Crosby, "The Measurement of Photoluminescence Quantum Yields. A Review," J PHYS CHEM, Vol. 75 (April 1971), p. 991.

Therefore $\sigma_{10}(\nu_f)\phi_0(\nu_f) = \eta(\tau_{spontaneous} \Delta\nu_f)^{-1}$. Because $\phi_0(\nu_f) = 8\pi n_d^2\nu_f^2/c^2$ we have

$$\sigma_{10}(\nu_f) = \frac{nc^2}{8\pi\tau_{spontaneous}(\Delta\nu_f)n_d^2\nu_f^2}$$

Then

$$\sigma_{10}(\nu) = \sigma_{10}(\nu_f) \frac{F(\nu)}{F(\nu_f)} \frac{\nu_f^2}{\nu^2}$$

Now $\sigma_{01}(\nu) = n^{-1} \ell^{-1} \ln T(\nu)$ where T is the transmission of a sample of concentration, n, and length ℓ. We may plot the ratio $\sigma_{01}(\nu)/\sigma_{10}(\nu)$. As shown in Equation 2c this is a crucial parameter in determining the threshold and tuning range of a dye laser and it often minimizes (i.e., minimum threshold power) far from the maximum of the emission band.